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EVALUATION AND MODELLING OF THE SPATIAL AND TEMPORAL VARIABILITY OF PARTICULATE MATTER IN URBAN AREAS

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ACADEMIC DISSERTATION in physics.

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Evaluation and modelling of the spatial and temporal variability of particulate matter in urban areas

Abstract

This thesis contains three subject areas concerning particulate matter in urban area air quality. Firstly, there is analysis of the measured concentrations of particulate matter mass concentrations in the Helsinki Metropolitan Area in different locations in relation to traffic sources, and at different times of year and day. Secondly, the evolution of traffic exhaust originated particulate matter number concentrations and sizes in local street scale are studied by a combination of a dispersion model and an aerosol process model. And thirdly, some situations of high particulate matter concentrations are analysed with regard to their meteorological origins, especially temperature inversion situations, in the Helsinki Metropolitan Area and three other European cities. The prediction of the occurrence of elevated particulate matter concentrations in the studied cities is examined, and the performance of current numerical weather forecasting models in the case of air pollution episode situations is considered.

The study of ambient measurement data revealed clear diurnal variation of the PM_{10} concentrations in the Helsinki Metropolitan Area sites, irrespective of the year and the season of the year. The diurnal variation of local vehicular traffic flows seemed to have no substantial correlation with the $PM_{2.5}$ concentrations, indicating that the PM_{10} concentrations were originated mainly from local vehicular traffic, while the $PM_{2.5}$ concentrations were mostly of regionally and long-range transported origin.
The modelling study of traffic exhaust dispersion and transformation showed that the number concentrations of particles originating from street traffic exhaust undergo a substantial change during the first tens of seconds after being emitted from the vehicle tailpipe. The dilution process was shown to dominate total number concentrations. Minimal effect of both condensation and coagulation was seen in the Aitken mode, but not in the accumulation mode number concentrations.
In the Helsinki Metropolitan Area, studied PM episodes were shown to be linked to predominantly stable atmospheric conditions with high atmospheric pressure and low wind speeds in conjunction with relatively low ambient temperatures. For the other European cities studied, the best meteorological predictors for the elevated concentrations of PM_{10} were shown to be temporal evolutions of temperature inversions, atmospheric stability and in some cases, wind speed. The studied weather prediction models were found to underpredict inversion magnitudes, and they overpredicted nocturnal ground surface temperatures and wind speed close to the ground. These tendencies would lead to underprediction of dilution and subsequently to underprediction of pollutant concentration levels.
The original papers are associated with various projects of the Atmospheric dispersion modelling group at the Air Quality Research Department of the Finnish Meteorological Institute.

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Nimeke

Kaupunki-ilman hiukkaspitoisuuksien ajallisen ja paikallisen vaihtelun arviointi ja mallinnus

Tiivistelmä

Tässä tutkimuksessa on tarkasteltu kaupunki-ilman hiukkasten pitoisuuksia kolmesta näkökulmasta. Aluksi tarkastellaan pääkaupunkiseudulla mitattuja hiukkasten massapitoisuuksia eri liikenneympäristöissä, ja pitoisuuksien vuodenaikais- ja päivittäisvaihtelua. Pakokaasun muuntumista ja laimenemista tarkastellaan katumittakaavassa lukumääräpitoisuuksien ja hiukkaskoon kannalta laimenemis- ja aerosoliprosessimallin yhdistelmän avulla. Lopuksi tarkastellaan, mitkä meteorologiset seikat vaikuttavat eräiden pääkaupunkiseudulla ilmenneiden korkeiden hiukkaspitoisuusjaksojen eli episodien syntyyn, ja vertaillaan neljässä Eurooppalaisessa kaupungissa ilmenneiden episodien hiukkaspitoisuuksia ja syitä. Käytössä olevien numeeristen säänennustusmallien kykyä ennustaa oikein sääolojen otollisuus hiukkasepisodin synnylle tarkastellaan.

Pääkaupunkiseudulla vuosina 1997-1999 tehtyjen hiukkasmittauksien perusteella havaittiin selvä päivittäinen hengitettävien hiukkasten- eli PM_{10}-pitoisuussykli riippumatta vuodenajasta ja tarkasteluvuodesta (arkipäivät). Pienhiukkasten eli $PM_{2,5}$-pitoisuuksien päivittäisvaihtelulla ei havaittu olevan merkittävää riippuvuutta liikennemääristä. Tästä päätellen PM_{10}-pitoisuudet olivat pääosin peräisin paikallisesta tieliikenteestä, ja $PM_{2,5}$-pitoisuuksiin vaikutti suurimmilta osin alueellinen- ja kaukokulkeuma.
Tieliikenteen pakokaasun laimenemisen ja muuntumisen tarkastelu aerosoliprosessin ja laimenemismallin yhdistelmällä osoitti, että pakokaasupäästöjen pienhiukkaset muuttuvat huomattavasti ensimmäisen muutaman kymmenen sekunnin aikana ympäristöön vapauduttuaan. Tutkituista aerosoliprosesseista laimeneminen osoittautui kyseisissä oloissa tärkeimmäksi hiukkasten lukumääräpitoisuuksiin vaikuttavaksi tekijäksi. Sekä kondensaatiolla että koagulaatiolla havaittiin minimaalinen vaikutus Aitkenmoodin lukumääräpitoisuuksiin, mutta akkumulaatiomoodin pitoisuuksissa vaikutusta ei ollut havaittavissa.
Ilmanlaadun hiukkaspitoisuusepisodien tarkasteluun oli valittu talvella tai keväällä tapahtuneita jaksoja, joiden syntyyn vaikuttivat ainakin osittain paikalliset tekijät. Tarkasteltujen pääkaupunkiseudulla havaittujen ilmanlaatuepisodien osoitettiin olleen yhteydessä olosuhteisiin, joissa vallitsi korkeapaine, tuulennopeudet ja lämpötilat olivat matalia ja ilmakehän stabiilius oli stabiili. Tutkittaessa kolmen muun Eurooppalaisen kaupungin tyypillisiä talvi/kevätepisodeja havaittiin, että parhaat kohonneiden PM_{10}-pitoisuuksien indikaattorit olivat vallitseva lämpötilainversio, ilmakehän stabiili stabiilius, ja joissakin tapauksissa myös matala tuulen nopeus. Ilmanlaadun hiukkasepisodin kehittymiselle otollisten sääolojen ennustamista tarkasteltiin usean säänennustusmallin tapauksessa yhden episodijakson aikana. Kaikkien mallien havaittiin aliarvioivan inversion voimakkuuden ja yliarvioivan inversiokerroksen korkeuden maanpinnasta, lämpötilan maanpinnassa sekä tuulen nopeuden lähellä maan pintaa. Episodinaikaista hiukkaspitoisuutta arvioitaessa nämä seikat johtaisivat laimenemisen yliarviointiin ja siten hiukkaspitoisuuksien aliarviointiin.

Tutkimusartikkelit liittyvät Ilmatieteen laitoksen Ilmanlaadun tutkimuksen mallimenetelmien tutkimusryhmän projekteihin.

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VI. Kukkonen, J., Pohjola, M., Sokhi, R.S., Luhana, L., Kitwiroon, N., Fragkou, L., Rantamäki, M., Berge, E., Odegaard, V., Slørdal, L.H., Denby, B., Finardi,S., 2005. Analysis and evaluation of local-scale PM_{10} air pollution episodes in four European cities: Oslo, Helsinki, London and Milan. Atmospheric Environment, Special issue: Fourth International Conference on Urban Air Quality: Measurement, Modelling and Management, 25-28 March 2003, edited by Ranjeet Sokhi, Vol 39/15, pp. 2759-2773.

1. Introduction

PM_x is a term used to denote the mass concentration (e.g. μg m^{-3} or ng m^{-3}) of aerosol particles suspended in the air. The size of the aerosol particles varies from nanometers to tens of micrometers in diameter, and a single particle can be composed of solid matter, liquid or be a mixture of solid and liquid constituents. The subscript x refers to the largest aerodynamic diameter included in the sample, aerodynamic diameter being defined as the diameter of a unit density (1 g cm^{-3}) sphere that has the same settling velocity as the particle, not considering its shape, density or physical size.

Generally, particulate matter of diameter less than 0.1 μm is referred to as "ultrafine particles, "fine particles" are of diameter less than 2.5 μm, and particles larger than 2.5 μm but smaller than 10 μm are referred to as "coarse particles" due to measurement purposes. Particulate matter can be characterised by several parameters including total mass or number concentration, mass or number size distribution and modality of size distribution. Usually different instruments are used to measure different particle characteristics in different size ranges (Morawska et al., 1999a).

Particulate matter mass concentrations, measured as PM_{10} and more importantly as $PM_{2.5}$, have been associated with hospital admissions and mortality in several studies conducted both in Europe and in the United States (Katsouyanni et al., 1997, Schwartz et al., 1996, Dockery and Pope, 1994, Pope et al., 1995). The World Health Organization evaluation done in 2003 suggests that for some health effects the fine fraction is more important and recommends the regulation of $PM_{2,5}$ instead of PM_{10}.

There is a strong indication that the adverse health effects of PM may not be due mainly to particle mass, but instead particle number concentration (Penttinen et al., 2001, Zhang et al, 2005) or the chemical composition of particles. Clearly, the particle size is a key factor regarding the effects of PM on human health, as the smaller the particle, the deeper it can penetrate into the human body via the lungs (Spurny, 1996, Siegmann et al., 1999). High number concentrations of fine and ultrafine particles in urban environments, especially in

the vicinity of major streets and roads (Buzorius et al., 1999; Morawska et al., 1999b; Pakkanen et al., 2001a), raise an interest to study the physical and chemical transformation of PM.

An air pollution episode can be defined as a situation during which air pollutant concentrations exceed a specified threshold value. In Finland, the limit values specified by the European Union comprise the set of applicable thresholds. For example, the hourly limit value for nitrogen oxide (NO_2) is 200 µg/m^3, allowing 18 exceedings per calendar year. For PM_{10} the hourly limit value is 50 µg/m^3 (allowing 35 exceedings per year) and 70 µg/m^3 over 24 hours, allowing an exceeding once in a month. The EU Directives require practical measures to be taken, if these air quality limits are exceeded. It should be noted that based on the studies included in the report, a limit below which the effects detrimental to human health would not exist could not be indicated (WHO, 2003).

Air pollution episodes depend on various factors including emissions, meteorological parameters, topography, atmospheric chemical and physical processes (including long range transport) and solar radiation. The relative importance of the various factors depends on the climatic characteristics, the geographical region and the season of the year. For example, particulate matter episodes in many cities occur in winter and spring times.

Finland as a Nordic country has its climate-dependent specific factors affecting the concentrations, chemical composition and sources of particulate matter. For example, springtime street-cleaning originated dust episodes, wintertime surface inversion promoted episodes, and wintertime cold starts and cold driving; these promote a Northern particulate matter phenomenology. Inversions, leading to stagnant air, are particularly important in relation to episodes in Finland and in many cases cause very high levels of pollution. In addition, regional and long range transport of pollution can also lead to elevated concentrations, e.g. for fine particulate matter.

Forecasting of episode situations is useful, because it enables national authorities to give warnings of impending high concentrations or to take precautionary measures for pollution control.

The general goal of this thesis was to add to the understanding of urban air quality issues concerning particulate matter by examination of the ambient PM concentrations in the Helsinki Metropolitan Area (HMA) and the factors affecting and leading up to situations of elevated ambient PM concentrations in the HMA. Specifically, the aims of this thesis were:

- to evaluate the concentration levels, sources and temporal variation and to a lesser degree, the spatial variation, of PM in the Helsinki Metropolitan Area (**Paper I**)
- to examine the importance of aerosol processes for the PM concentrations near a traffic source (**Papers II, III**)
- to find out which meteorological variables are significant in the development of elevated PM concentrations and can be used to predict the occurrence PM episodes in the Helsinki Metropolitan Area in the local scale, and to evaluate the performance of current numerical weather forecasting models in the case of air pollution episode situations (**Papers IV,V,VI**)
- to compare and explain typical air pollution episode conditions in some European cities (**Paper VI**).

2. Review of literature

2.1 Particulate matter in the urban environment

According to Seinfeld and Pandis (1998), sources of particulate matter in the air of urban environments include vehicular traffic, industrial sources, construction sites, energy production, vegetation (pollen, debris), suspension of mineral dust from streets and other surfaces by wind induced turbulence, and regional and long-range-transported particulate material. Traffic induced particulate matter contains exhaust particles, particles produced by tire wear and studded tires, and particles suspended from the street surface by traffic-induced turbulence.

The processes affecting particulate matter in the atmosphere are homogeneous nucleation, condensation/evaporation, coagulation, dilution, wet and dry deposition and suspension.

Homogeneous nucleation is the process where liquid droplets are formed from vapours in the ambient gas phase, consisting then only of a liquid phase formed by the vapour molecules. By condensation it is meant that gas molecules are transported to existing surfaces (foreign nuclei or other surfaces) by diffusion, and evaporation is removal of molecules from the particle surface liquid phase into the gas phase. Condensation is dependent on the vapour pressure gradient between the particle surface and ambient air, particle size, mass diffusion and conduction of heat. Condensation changes the particle size and chemical composition, but not the number concentration of the particles. In the coagulation process particles collide due to their motion in air (Brownian motion, differences in gravitational settling velocities, turbulent flows, external force fields) and then stick to each other. Coagulation is least efficient between particles of same size, and its overall influence decreases with increasing particle size. Coagulation affects both particle number concentration and size and may affect particle composition. Wet and dry deposition are particle removal processes. In dry deposition the particles are transported to surfaces (ground, other available surfaces) where they stick. Wet deposition entails the interaction of the particles with falling drops of rain, snowflakes, or settling fog droplets. Suspension

means the subsequent lifting of deposited particles from surfaces back into the atmosphere by turbulence (wind or e.g. traffic induced).

By looking at the generation processes, fine particles can be divided roughly into three size modes: the nucleation, Aitken and accumulation modes. The nucleation mode, of diameters between approximately 1 to 20 nm, is formed by nucleation and condensation of vapors from combustion processes and nucleation of compounds in the atmosphere and subsequent condensation of vapors. Part of nucleation mode particles are so-called primary particles, formed before emission from vehicle engines (Laakso et al., 2003). The majority of the Aitken and accumulation mode particles in urban environments are primary particles. Secondary Aitken mode particles are formed when nucleation mode particles grow by condensation and coagulation to diameters of typically 20 – 100 nm. Further growth of Aitken mode particles by the same processes as well as by cloud processing forms accumulation mode particles of diameters 0.1 – 1 μm. Accumulation mode particles are also emitted from industrial processes, suspension from road beds, and sea spraying. Coagulation among accumulation mode particles is too slow to be of importance in the formation of coarse mode particles, which are therefore originated from dust suspended from surfaces by wind and turbulence caused by vehicles, from emissions from vehicles and industry, sea spray, plant particles and long-range transported materials.

2.1.1 Field observations of particulate matter

Measured urban PM_{10} and $PM_{2.5}$ concentrations and their spatial and temporal variations in European urban locations have been discussed widely by, e.g., Monn et al. (1995) for Switzerland, Harrison et al. (1997) and Kingham et al. (2000) for UK, Johansson et al. (1999a) for Sweden, Ojanen et al. (1998) for Finland and Berner et al. (2004) and Gomišček et al.(2004) for Austria, just to mention a few. However, the amount of data and the time periods considered in the above-mentioned studies have been fairly limited. In some studies carried out in North America, more extensive time periods have been addressed (Brook et al., 1999, and Darlington et al., 1997, Lall et al., 2004), and in Europe by van Dingenen et al. (2004) and Querol et al., (2004). Some studies have addressed

systematically the influence of the relevant meteorological parameters on urban PM concentrations; e.g. Monn et al. (1995), Chu (2004) and Buchanan et al. (2002).

When considering number concentrations of particulate matter, generally in urban environments the total particle concentration in the ambient air can be of the order of 10^4-10^6 cm^{-3} depending on distance to emission sources, most of the particles being smaller than 0.1 μm (Seinfeld and Pandis, 1998). Buzorius et al. (1999) have analysed PM number concentrations (from 10 to 500 nm in particle diameter) in the city of Helsinki, and reported measured peak number concentrations of the range of 80 000 cm^{-3} (daily average) and 10 minute average concentrations of range of 10^3 cm^{-3} in regional background site and 10^5 cm^{-3} for downtown traffic sites, for the years 1996-1997. Hussein et al. (2005) reported for Helsinki highest number concentrations of the order of 60 000 cm^{-3} for winter season in an urban traffic environment. Ketzel et al. (2004a) reported average measurement values of total particle numbers of 7700 cm^{-3} for Copenhagen in autumn to winter 2002. Particle number concentration measurements as function of distance to road or street have been reported by, e.g., Hitchins et al. (2000), Zhu et al. (2002a), Zhang et al. (2004), Rosenbohm et al. (2005) and Pirjola et al. (2006).

Both particle mass concentration and number concentration can be used in examination of particulate matter. When comparing the diameters where peak concentration values are found in vehicle exhaust and urban ambient air measurement data, the mass concentration peak diameters do not correspond with the diameters at which the number concentration peaks are found. The mass of a small number of large particles can be significantly larger than the mass of a large number of small particles. Morawska et al. (1999a) found a linear relation between the mass and number concentration of submicrometer aerosol measured at the same location during summer. Harrison et al. (1999) found significant linear correlation between particle number concentrations and $PM_{2,5}$ and PM_{10} for an urban background location in Birmingham, UK. In highly polluted environments, as reported by Mönkkönen et al. (2004) for PM_{10} in New Delhi, India, and Longley et al., (2003) for Manchester, UK, the relationship seems not to be linear, as high mass concentration of particulate matter provides a large coagulation sink.

Ketzel et al. (2004a) have studied the contribution of urban versus regional sources of total particle number and PM_{10} from measurement data with simultaneous measurement of particle size distribution and PM_{10} at one urban, one near-city and one rural location in Scandinavia. They found that the traffic source contributes strongest in the size range of 10-200 nm, and that maximum of the number size distribution was at 20-30 nm at kerbside and at 50-60 nm at rural level. Giugliano et al. (2005) have studied the effect of the traffic source concerning PM_1, $PM_{2,5}$ and PM_{10} at urban sites with various traffic situations within the city of Milan in Italy. Forsberg et al. (2005) estimated the local contribution to PM_{10} to be about 29% in Southern urban environments in Sweden and as much as 50% in the Northern parts due to diminishing importance of long distance transport.

In addition to ambient concentration measurements and size distribution and chemical composition measurements, another view into the traffic source, necessary for PM modelling, is emission measurement of the same parameters. Emission factors derived for particulate matter are defined as mass or number of particles emitted per kilometer driven (g/km of particle number/km) or per unit of mechanical energy delivered (g/kWh), per vehicle (Morawska et al. 2005). The majority of emission factor studies published have been on total mass emissions, but studies have also been published on particle number emission factors for motor vehicles. Vehicular emission factors can be determined by chassis dynamometer measurements (e.g. Norbeck et al., 1998, Lehmann et al., 2003), indirect estimations based on measurements near roads (e.g. Gramotnev et al., 2003, Morawska et al., 2005, Ketzel et al., 2003), or by chasing experiments by mobile measurement laboratories (Kittelson et al., 2004, Yli-Tuomi et al., 2005, Pirjola et al., 2006). Emission factor studies have also been done for non-exhaust emissions including tyre and road surface wear and traction sanding of the roads (Dahl et al., 2006, Kupiainen et al., 2005) and suspension of materials deposited on nearby surfaces (Omstedt et al., 2005).

2.2 Aerosol process modelling studies

At this time, some aerosol process models that can be used in evaluating urban scale atmospheric transformation of PM when combined with models describing the dilution

from the urban sources, have been published in the literature. Such aerosol process models are required to include aerosol dynamic processes in sufficient detail, such as nucleation, condensation and evaporation, coagulation and deposition. They should also be sufficiently efficient computationally. Some of the models that have been used in studies of aerosol transformation processes in North European urban surroundings are reviewed next.

The AEROFOR2 model (Pirjola, 1999; Pirjola and Kulmala, 2001) is a Lagrangian type sectional box model; it is assumed that the particles can consist of soluble, weakly soluble and insoluble material, and the particle population can be externally or internally mixed. The model contains emission of gases and particles, gas-phase chemistry, binary homogeneous nucleation of sulphuric acid and water, ternary homogeneous nucleation of sulphuric acid, ammonia and water, multicomponent condensation (sulphuric acid, ammonia, water and generic organic vapour) onto pre-existing particles, SO_2 uptake by particles, inter- and intramode coagulation of particles deposition of particles, and dilution and mixing with ambient air.

Vignati et al. (1999) have evaluated the relative importance of various aerosol processes on the number concentrations of PM in urban environments. They combined a vehicular plume model with a simplified particle transformation model to assess the impacts of coagulation, condensation of water vapour and plume dilution on the size distribution of particles in selected urban environmental conditions. However, this study did not consider explicitly the influence on the computed results of the chemical composition of the particles. AERO3, reported by Vignati (1999), is a box aerosol dynamics model, which describes the coagulation, condensation and nucleation of three particle populations consisting of sulphuric acid-water droplets, black carbon, and particles which are a mixture of sulphuric acid, water and black carbon. The particle size range of 0,001 - 31,6 μm is described by 46 size classes.

Wehner et al. (2002) used a sectional aerosol dynamics model to simulate transformation and dilution in a street canyon in a German urban area. The processes included were

condensation of H_2SO_4 and coagulation. They reported that the simulation time to reach the urban background particle size distribution was 15 to 50 minutes.

Gidhagen et al. have coupled the aerosol dynamics model MONO32 (Pirjola and Kulmala, 2000, Pirjola et al., 2003) (see chapter 3.2 concerning the modelling methods used in this thesis) with the finite volume based computational fluid dynamics model StarCD for the simulation of evolution of the size distribution in several microenvironments in Sweden, e.g. road tunnel (2003), street canyon (2004a) and the vicinity of a highway (2004b). For the study of particle number concentrations in a larger, urban area scale, Gidhagen et al. (2005) used a combination of MONO32 and the dispersion model MATCH (Multi-scale Atmospheric Transport and Chemistry model). In these studies the aerosol transformation processes included were coagulation and deposition, but condensation and nucleation were not considered. The effect of aerosol processes in reducing the total number concentrations was reported to be more important in environments and situations where particle number concentrations were significantly higher than in a "normal" highway environment, e.g. in road tunnels and during peak episodes.

Ketzel et al. (2004b) used the AERO3 aerosol dynamics model by Vignati (1999) in the Multi-plume Aerosol dynamics and Transport model (MAT) where dilution was described by a combination of a plume model with a 1D Lagrangian trajectory framework in an urban scale. Dilution, deposition, coagulation and condensation were included in the model system, and chemical composition of the aerosol was described with only one component, while 121 size classes were used for the particle diameter range of 2 nm to 2 μm. Condensing species were H_2SO_4 and low volatile organics, for which the properties of H_2SO_4 were used. During the studied time scale of 12000 s the considered particle dynamics processes were found to act mainly at the lower end of the size spectrum, while only small changes appear for particle sizes larger than 100 nm.

Considering theoretical analysis of the importance of aerosol processes in urban environments, Zhang and Wexler (2002) have analysed the relative importance of different aerosol processes in ambient air and in their later analysis (Zhang and Wexler, 2004, Zhang

et al., 2004)) they separated two stages for dilution near roadways, the tailpipe-to-road stage where dilution is induced by traffic generated turbulence in the first 1 – 3 s, and the road-to-ambient stage lasting about 3 – 10 min. Ketzel et al. (2004c) have also reported on analysis of time scales of aerosol processes in UFP aerosol of diameter < 0,1 μm; the sizes which are characteristic for aerosol emitted from vehicle exhaust pipe) during dilution at kerbside and on the urban level.

2.3 Particulate matter episodes

Air pollution episodes may be of local, regional and long-range transported, or of mixed local and regional and long-range transported origin. Local episode sources in urban environments include vehicular traffic, domestic heating, local fires, construction sites and industry including energy production. Regional and long-range transported episode sources include forest fires, agricultural fires, and distant urban or industrial centers.

The work of QUARG (1993, 1996) and Railo (1997) has shown that under episodic conditions, concentrations of air pollutants can considerably exceed national and international standards and limit values. European peak pollution episodes have been reviewed within the COST 715 action "Meteorology applied to Urban Air Pollution Problems" (e.g., Fisher et al., 2001, Kukkonen, 2001). The formation and duration of air pollution episodes depend on various factors including emissions, local and synoptic scale meteorological conditions, topography, and atmospheric chemical processes. Sokhi et al. (2002) and Piringer and Kukkonen (2002) have shown that the relative importance of such factors is dependent on the geographical region, its surrounding emission source areas and the related climatic characteristics, as well as the season of the year.

Analyses and evaluation of selected historic local episodes have been previously published for some European cities, including Oslo (Berge et al., 2002a), Helsinki (Mäkelä et al., 1998, Berge et al., 2002a, Karppinen et al., 2002), London (Sokhi et al., 2002) and various Northern Italian cities (Finardi, 2002, Sokhi et al., 2002). Examples of episodes of PM_{10}, NO_2 and O_3 were presented and the causes were examined in relation to local emissions and meteorological conditions. For both particulate matter and nitrogen oxides, low lying

inversion and local low wind speeds were shown to be particularly important in that they tend to lead to high concentrations of air pollutants.

Regional and long-range transport of pollution can also lead to limit values being exceeded, for example for fine particulate matter. In Nordic countries, atmospheric long-range transport constitutes an important part of the total urban background $PM_{2.5}$ concentration (e.g., Johansson et al., 1999b; Pakkanen et al., 2001b,c). Karppinen et al. (2004) estimated the contribution of long-range transportation to the $PM_{2,5}$ concentrations measured in Helsinki to be 64 – 76% for the years 1998-2000. Ketzel at al. (2004a) reported on three episodes of elevated PM_{10} during late autumn 2002 in Copenhagen. These elevated concentrations were indicated to be of long-range transported origin. The meteorological analysis contained only wind direction and wind speed data.

Watson and Chow (2002) analysed a wintertime $PM_{2.5}$ episode that occurred in California's San Joaquin Valley in January 2000. The episode lasted for approximately ten days. The influence of regionally and long-range transported $PM_{2.5}$ was substantial, as half or more of the urban fine particulate matter concentrations were present at surrounding non-urban locations. Primary particles accumulated during early morning and nighttime, decreasing when a shallow radiation inversion coupled to a valleywide air layer. Liu and Chan (2002a,b) analysed a two-day episode with increased concentrations of NO_x, RSP (Respirable Suspended Particulates) and SO_2 in Hong Kong in December 1999. They concluded that local vehicular and stationary emissions were mainly responsible for the increased concentrations. The stably stratified synoptic conditions combined with sea-land breezes over a complex topography were reported as the main meteorological factors affecting the concentrations.

Piringer and Kukkonen (2002) have shown that inversions, which lead to stagnant air, are particularly important in relation to episodes and these are in many cases responsible for very high levels of pollution. Although inversions are common in Finland, strong surface inversions with strengths of more than 10˚C occur only in wintertime (Huovila et al., 1991, Karppinen et al., 2001). The strength of a surface inversion is defined as the temperature

difference between the top of the inversion and the ground. Accuracy in forecasting air pollution episodes therefore depends crucially on the ability of the numerical weather prediction models to forecast the strength of these inversions.

For cities in northern climates, the coarse particle fraction is important for the elevated PM_{10} concentrations especially in spring (e.g., Johansson, 1999a, Vallius et al., 2000, Kuhns et al., 2003) These so-called springtime dust episodes originate in the use of sand, salt and studded tyres to increase traction on icy streets. Mineral dust from the sand and pavement, tyre and stud material and deposited particles (including exhaust and other) can be lifted into the air by mechanical disturbances, such as wind and traffic-induced turbulence, when the road surface is freed from snow and ice and dries up in the spring.

3. Data and methods used in this work

3.1 Studied areas, time periods and measurement equipment

This thesis is largely based on measurement data collected firstly by the Helsinki Metropolitan Area Council air quality monitoring network (years 1995, 1997-1999 and 2002), presented in Figure 1, and secondly by the network of meteorological measurement sites in the same area.

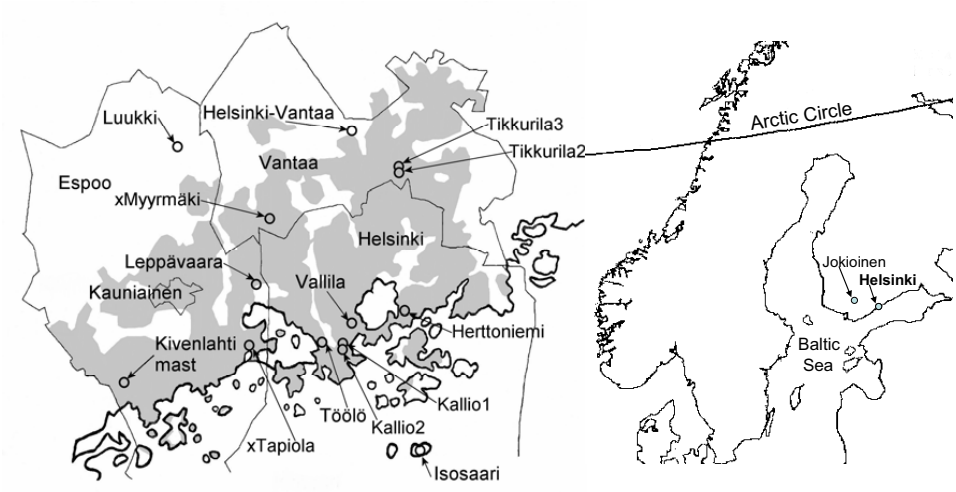


Figure 1. The locations of the air quality monitoring network measurement sites and meteorological measurement sites within the Helsinki Metropolitan Area, comprising four cities (Helsinki, Espoo, Vantaa and Kauniainen) during the years studied in this thesis. The size of the depicted area is approximately 35 x 25 km^2. The areas with the highest proportion of buildings (vs. other forms of land use) have been presented with grey shading. The stations marked with "x" were moved elsewhere after measurement campaign duration of one year.

The air quality measurement data was from the locations of Vallila, Töölö, and Kallio2 in central Helsinki, Herttoniemi in urban Helsinki, Tikkurila and Myyrmäki in Vantaa, and Leppävaara, Luukki and Tapiola in Espoo. The meteorological measurements were collected at the sites Kivenlahti (radio tower), Helsinki-Vantaa airport, Isosaari and Kallio1. In addition to the studies on the Helsinki Metropolitan Area (HMA), **Paper VI** contains

studies of 3 other cities in Norway, Great Britain and Italy. Table 1 contains a summary of the measurement data (locations, time periods) included in this work. HMA air quality measurement network site classifications are presented in Table 2. A summary of the measurement methods and associated size ranges (PM_1, $PM_{2,5}$, PM_{10}) is presented in Table 3.

Table 1. Measurement locations and time periods included in this thesis.

Paper	Geographical area	Number of air quality stations	Number of meteorological stations	Time period
I	HMA	5	2	1997 - 1999
II-III	Urban boreal	-	-	Midday, summer
IV-V	HMA	4	3	27-29 Dec 1995
VI	HMA Oslo, Norway London, Great Britain Milan, Italy	4 6 7 3	3 3 3 1	3-14 Apr 2002 4-1 Jan 2003 18-27 Feb 2003 14-19 Dec 1998

Table 2. The measurement site type classifications of the air quality measurement network sites.

Measurement site	Site type classification	Distance to nearest traffic source, m
Vallila	Urban traffic	14
Töölö	Urban traffic	<2
Kallio 2	Urban background	80
Herttoniemi	Suburban traffic, mobile	50
Tikkurila	Urban	7
Myyrmäki	Suburban traffic, mobile	15
Leppävaara	Suburban traffic	15
Luukki	Regional background	800
Tapiola	Suburban traffic, mobile	50

Table 3. Summary of particulate matter species and concentration measurement methods.

Paper	Measured quantity	Measurement method
I	PM_{10} $PM_{2,5}$	TEOM Eberline FH 62 I-R
II-III	PM_1	Literature reference
IV-V	PM_{10}	TEOM
VI	HMA: PM_{10} HMA: $PM_{2,5}$ Oslo: PM_{10} Oslo: $PM_{2,5}$ London: PM_{10} Milan: PM_{10}	TEOM, Eberline FH 62 I-R Eberline FH 62 I-R TEOM, Eberline FH 62 I-R Eberline FH 62 I-R, Partisol 2025 TEOM TEOM

3.2 The numerical models

A summary of the numerical models used in this thesis is presented in Table 4. The models used can be categorized into meteorological preprocessing models, aerosol process models, models describing dispersion, and numerical weather forecasting models.

Table 4. Summary of the models used.

Paper	Model name	Model type	Modeled output variables used in the analyses	Reference
I	MPP-FMI	Meteorological pre-processing	Monin-Obukhov length scale, boundary layer height	Karppinen et al., 1998, 2000a.
II-III	MONO32	Aerosol process model	Particle population section number concentration, chemical composition, radius	Pirjola and Kulmala, 2000, 2001. Pirjola et al., 2003.
	(unnamed)	Plume dispersion model	Dilution of particle number concentration	Vignati et al., 1999.
IV	HIRLAM Version 4.6.2	Numerical weather forecasting model	Wind speed and direction, temperature, relative humidity	http://www.knmi.nl./hirlam/
	MM5	Numerical weather forecasting model	Temperature	Dudhia, 1993. Berge et al., 2002b.
	MPP-FMI	Meteorological pre-processing	Monin-Obukhov length scale, boundary layer height	Karppinen et al., 1998, 2000a.
	Urban Dispersion Modelling System (FMI)	Dispersion modelling system	Emissions, spatial and temporal dispersion of concentrations	Karppinen et al. 2000b.
V	HIRLAM Versions 4.6.2 and 6.2.1	Numerical weather forecasting model	Temperature, wind speed, geostrophic vorticity	Eerola, 2000, Järvenoja, 2004.
VI	HIRLAM 4.6.2	Numerical weather	Synoptic meteorological	http://www.knmi.nl./hirlam/

		forecasting model	situations for Helsinki and Oslo	
	ECMWF Cycle25r1	Numerical weather forecasting model	Synoptic meteorological situations for London and Milan	http://www.ecmwf.int/research/ifs docs/
	MM5	Numerical weather forecasting model	Meso-and microscale meteorological for London	Dudhia, 1993.
	MPP-FMI	Meteorological pre-processing	Monin-Obukhov length scale for Helsinki	Karppinen et al., 1998, 2000a.
	SURFPRO	Meteorological pre-processing	Monin-Obukhov length scale for Milan	Finardi et al., 1997.

3.2.1 Meteorological pre-processing models

MPP-FMI is a meteorological pre-processing model adapted specifically for urban environments (Karppinen et al., 1998 and 2000a). The model is based mainly on the energy budget method of van Ulden and Holtslag (1985). The model utilises meteorological synoptic and sounding observations, and its output consists of hourly time series of relevant atmospheric turbulence parameters (the Monin-Obukhov length scale, the friction velocity and the convective velocity scale) and the boundary layer height.

SURFPRO is a meteorological pre-processing model (Finardi et al., 1997 and Arianet, 2002) also based on the energy budget method of van Ulden and Holtslag (1985), and the work of Paine (1988). The model utilises surface based data and vertical profiles of temperature, and evaluates atmospheric turbulence scaling parameters and the boundary layer height.

3.2.2 Aerosol process model

MONO32 by Pirjola and Kulmala (2000) is a simplified version of the Lagrangian aerosol process model MULTIMONO. The term ‘MULTI’ refers to multicomponent condensation of different vapours, and the term ‘MONO’ indicates that each size section is assumed to be

monodisperse. The model includes gas-phase chemistry and aerosol dynamical processes. Gas-phase chemistry is based on the EMEP mechanism (Simpson, 1992). Particles were classified into four size fractions in this work, the initial diameters could vary between 1 and 20 nm in the nucleation mode, 20 – 100 nm in the Aitken mode, 100 nm – 2.5 μm in the accumulation mode, and are larger than 2.5 μm in the coarse mode. In the MONO32 model, all particles in a certain size section are represented as to have the same chemical composition. This can be a mixture of water, sulphuric acid, ammonium nitrate and ammonium sulphate, organic and elemental carbon, sea salt (NaCl) and mineral dust. The aerosol processes included in the model at the time were homogeneous binary sulphuric acid–water nucleation and ternary sulphuric acid-water-ammonia nucleation, multicomponent condensation of sulphuric acid and organic vapour, inter- and intramode coagulation, and dry deposition (Pirjola et al., 2003).

3.2.3 Dispersion models

Plume model by Vignati et al. (1999) is a simple jet and plume model, which allows for the initial momentum of the exhaust gases (turbulent jet), and the influence of street-level wind and traffic-induced turbulence, which is calculated according to Berkowicz et al. (1997). The model is applicable in the description of dispersion in the immediate vicinity of the emission sources near the ground level; at distances smaller than approximately 100 m from source.

Urban Dispersion Modelling System reported by Karppinen et al. (2000b) contains dispersion and emission modelling for stationary sources by the model UDM-FMI (Urban Dispersion Model, a multiple source Gaussian plume model taking into account point, area and volume sources)) and dispersion modelling of vehicular traffic sources by model CAR-FMI (Contaminants in the Air from a Road; road network Gaussian line source dispersion model) (Härkönen, 2002), estimation of traffic flows (traffic volumes and traveling speeds) by the transportation planning system EMME/2 (INRO, 1994) and emissions and vehicular sources by the LIISA model (Mäkelä et al., 1996), the meteorological pre-processing model MPP-FMI, chemical transformation models and the statistical and graphical analysis of the computed times series of concentrations.

3.2.4 Numerical weather forecasting models

HIRLAM version 4.6.2 was the operational numerical weather forecasting model in Finnish Meteorological Institute from November 1999 to March 2003, (Eerola 2000). It is a hydrostatic grid model that can be executed with two horizontal resolutions, 22 km and 44 km, for the synoptic and regional scale HIRLAM versions, respectively. The synoptic scale resolution was used in **Paper IV**. The larger computational domain covers Europe and the Northern Atlantic, and the smaller one Northern Europe, the British Isles, and part of continental Central Europe and Russia. In both model versions, the number of horizontal grid points is 194 × 140, and there are 31 vertical grid levels. The boundary values for the larger domain are taken every six hours from the global ECMWF-model, and the smaller domain boundary values are taken from the larger domain every three hours. The model output includes the following data for 10 standard pressure levels between 1000 and 70 hPa: wind, temperature, relative humidity, geopotential height and vertical velocity. The model also evaluates surface pressure and temperature, accumulated convective and stratiform precipitation and their intensities, sea surface temperature, ice coverage and albedo. HIRLAM version 6.2.1, operational since February 2004, has 40 vertical grid levels and 22 km horizontal resolution (number of grid points is 438 × 336), semi-Lagrangian time integration instead of the former Eulerian and changes in surface analysis and parameterization among other improvements.

ECMWF model is the **"**European Center for Medium-range Weather Forecasts" model, reported in http://www.ecmwf.int/research/ifsdocs/.

MM5 is a non-hydrostatic numerical weather forecasting model (Fifth Generation NCAR/Penn State Meso-Scale Modeling System) (Dudhia, 1993, Berge et al., 2002b). In this thesis MM5 was used with set-ups of three nests with the horizontal resolution of 9, 3 and 1 km in **Paper IV** and five nests with the horizontal resolution of 81, 27, 9, 3 and 1 km in **Paper VI**. The meteorological input data at the outer domain boundaries used for the simulations was obtained from the ECMWF model computations.

4. Results and discussion

4.1 Diurnal and spatial variations of particulate matter concentration levels in the Helsinki Metropolitan Area

According to the compilation of air quality measurement data, made for the years 1997-1999 (presented in **Paper I**), the hourly average PM_{10} concentrations at the urban traffic type measurement site most resembling kerbside conditions, Töölö, during working days varied between the maximum values of approximately 40 µg m^{-3} during the morning rush hour at 8 a.m. and nighttime values of about 18 µg m^{-3}, while the $PM_{2,5}$ concentration values varied between about 8 µg m^{-3} and 13 µg m^{-3}. Querol et al., (2004) reported values of 26-51 µg m^{-3} and 13-18 µg m^{-3} for kerbside type sites for Sweden and 30 – 55 µg m^{-3} and 20 - 39 µg m^{-3} for other European kerbside location PM_{10} and $PM_{2,5}$ concentrations, respectively.

At the urban background site (Kallio2) the hourly average $PM_{2,5}$ concentration was about 10 µg m^{-3}, similar to the range of 8 - 15 µg m^{-3} reported by Querol et al. (2004) for Swedish corresponding sites; both are substantially lower than the values they report for other European sites (15 - 30 µg m^{-3}).

Van Dingenen et al. (2004) reported European continental background concentration values of 7 µg m^{-3} (±4,1) and 4,8 µg m^{-3} (±2,4) for PM_{10} and $PM_{2,5}$ respectively, based on a synthesis of data from 31 measurement sites in Europe (eastern Europe not included) and the years 1998-2000. The study of 21 European cities' measurement site $PM_{2,5}$ data from the years 2000-2001 by Hazenkamp-von Arx et al. (2004) reported annual mean background values ranging from 3,7 (Reykjavik, Iceland) to 44,9 µg m^{-3} (in Turin, Italy). Compared to the Hazenkamp-von Arx study, for the year 1999, the urban background $PM_{2,5}$ concentration for the Helsinki station was at the lower end of the range with about 10 µg m^{-3}, and the PM_{10} concentration the regional background value at Luukki was 10 µg m^{-3}.

The diurnal variation of the PM_{10} concentrations in the Helsinki Metropolitan Area urban and suburban type measurement sites was clear, irrespective of the year and the season of the year. An example of the PM_{10} variation during the year 1999 is presented in Figure 2a. This

variation partly follows the corresponding variation of local vehicular traffic flows, with discrepancy suspected to be caused by suspension of PM from street surfaces.

During working days, there is a very clear diurnal variation of local vehicular traffic. Both the spatial and temporal variations of the fine particle ($PM_{2.5}$) concentrations were moderate, except for a moderate increase during the morning rush hours as presented in the Figure 2b for the example year 1999. Therefore the diurnal variation of local vehicular traffic flows seems not correlated with the $PM_{2.5}$ concentrations.

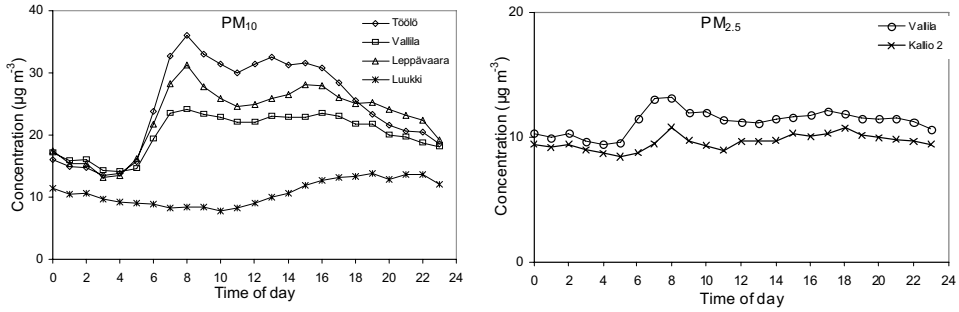


Figure 2. The diurnal variation of PM concentrations during working days in 1999. a) PM_{10} concentrations in the urban air quality measurement stations of Töölö, Vallila, and Leppävaara and the urban background station Luukki. b) $PM_{2,5}$ concentrations in the urban air quality measurement stations of Vallila and Kallio2 (urban background).

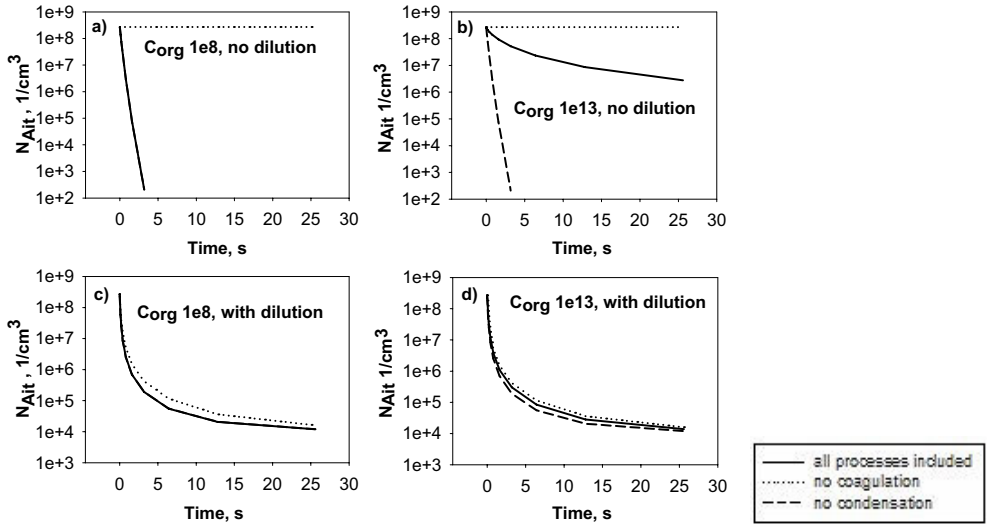
The results provide indirect evidence indicating that the PM_{10} concentrations are originated to a large extent from local vehicular traffic (direct emissions and suspension), while the $PM_{2.5}$ concentrations are more of regionally and long-range transported origin. This result is qualitatively in agreement with source apportionment studies in the same area by Ojanen et al., (1998), who evaluated approximately 40 % of the $PM_{2.5}$ concentration to be originated from local sources, and the rest of the $PM_{2.5}$ mass from regional or long-range transported pollution. More recently, Karppinen et al. (2005) estimated the contribution from regional and long range transported origins to be less than 50% in Helsinki centre (and nearly 100% in the outskirts of the Helsinki Metropolitan Area). Querol et al. (2004) have reported kerbside European traffic contributions of 35 – 55% for PM_{10} and 40-60% contribution for $PM_{2,5}$.

4.2 Aerosol process modelling in street scale

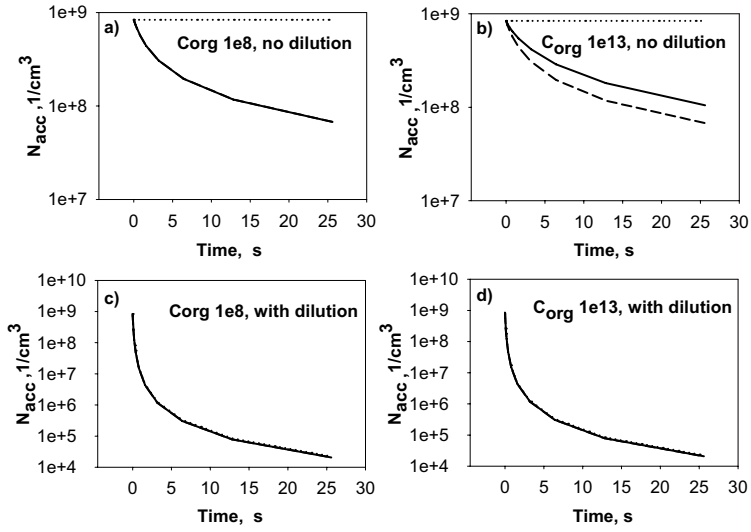
A modelling study, reported in the **Papers II and III,** was made in order to examine the importance of aerosol processes on fine particles from traffic exhaust in local scale in street environment. The vehicle exhaust particle size distribution used was composed from Lappi et al. (2000) for light-duty diesel vehicles. These distributions correspond to a driving speed of 55 km/h, considered a typical value in the Helsinki Metropolitan Area. The chemical composition of various size classes was evaluated based on Kittelson et al. (1999) and for simplicity, the particles were assumed to consist of 20 % organic carbon and 80 % elemental carbon. The particle number concentrations for background urban air correspond to values that are characteristic for the Helsinki Metropolitan Area, measured in Helsinki by Koponen et al. (2001) for the urban background. For the urban background a composition from the work of Viidanoja et al., (2002) was used. The condensable vapours considered were sulphuric acid, a generic organic vapour, e.g. nonadecane (Abdul-Khalek et al., 2000), and water. The modelling input values were chosen to represent midday summer conditions with wind speed 3 m s^{-1}**.** This value was chosen as it was reported as a characteristic annual average wind speed for HMA by Kukkonen et al. (2001).

The effects of coagulation, condensation and plume dilution on the evolution of the assumed vehicular exhaust aerosol, concerning particle number concentrations, radii and chemical composition, were considered separately for each particle size mode. The effect of each specific process was studied by including all other processes and neglecting one process at a time in the model computations.

As examples of the type of modelling results obtained, the time evolution of the predicted total number concentration is presented in Figure 3, the predicted number concentrations in the accumulation mode in Figure 4, and the particle radii of the Aitken mode in Figure 5. The figures presented represent examples of low and high values of C_{org}, and do not include the results of predictions that were calculated with in-between values of C_{org}.



Figures 3 a-d. The total number concentration of Aitken mode particles against time. The left-hand and right-hand-side sets of figures correspond to the cases where the concentration of the organic condensable vapour is 10^8 (molecules) cm^{-3} and 10^{13} cm^{-3}, respectively. The legend shows which processes have been included in the computations. The corresponding distance scale in all figures is approximately 75 m.



Figures 4 a-d. The total number concentration of accumulation mode particles against time. The notation is the same as in Figure 3.

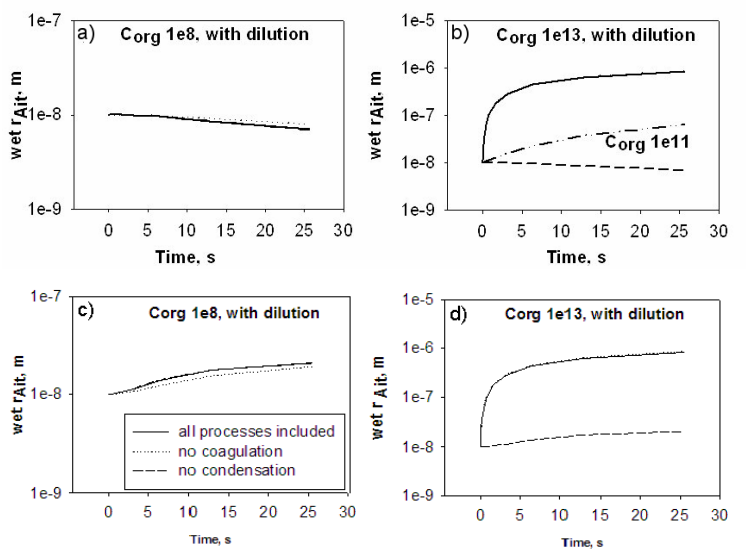


Figure 5 a-d. The evolution of the wet radii of the Aitken mode particles in cases with dilution included. A-b) Original results from **Paper II**. c-d) The corrected results from **Paper III**. The left-hand and right-hand-side sets of figures correspond to the cases where the concentration of the organic condensable vapour is 10^8 (molecules) cm^{-3} and 10^{13} cm^{-3}, respectively. The legend shows which processes have been included in the computations. The corresponding distance scale in all figures is approximately 75 m.

The results indicated that for the ambient conditions (wind speed, temperature) chosen for this case, condensation of an insoluble organic vapour is important under the selected conditions, if its concentration exceeds a threshold value of 10^{10} or 10^{11} cm^{-3} (not presented in the figures) for the Aitken and accumulation mode particles, respectively. The condensation or evaporation of water can also be important processes; however, its influence is strongly dependent on the hygroscopicity of particles. The effect of coagulation is substantial only, if the dilution of the exhaust plume is neglected.

With the ambient conditions studied in this work, the number concentrations of particles originating from street traffic exhaust underwent a substantial change during the first 25 s after being emitted from the vehicle tailpipe. Considering particle measurements in traffic

type sites this indicates that the measurement devices' distance from the traffic emission source would have a "turning point" value, from where closer to the source the measurements can be expected to vary substantially. This distance would depend on wind direction, wind speed and atmospheric stability (vertical transport).

This work did not consider possible nucleation processes, and evaporation neither. Nucleation was supposed to have already happened and the particles produced by it were included in the initial emission.

The results of Vignati et al. (1999) concerning the relatively small effect of coagulation and condensation compared to dilution are similar with the results obtained in **Papers II and III**. The particle number concentration measurement data available to Vignati et al. was considerably better than the PM mass data available for the evaluation of the modelling in **Papers II and III**.

Of the work published by Gidhagen et al., the highway vicinity study (2004b) is closest to the conditions in the **Papers II and III**. Gidhagen stated that to see significant coagulation reductions in total number concentrations (the order of 10%,) the emitted particles' evolution should be simulated for significantly longer distance than a hundred meters; this result is similar to the findings in **Papers II and III**. On the other hand, Zhu et al. (2002a,b) concluded from their studies, that both atmospheric dispersion and coagulation contribute to the rapid decrease of particle number concentration near roads.

4.3 Particulate matter air pollution episodes in Helsinki Metropolitan Area and other European cities

In this thesis, several air pollution episodes containing significant elevations of PM concentrations have been studied. The included episodes were chosen on the basis of occurrence in either winter or spring, and having at least partly local origin.

4.3.1 Episode in March 1998 in the HMA

The spring PM episode considered in **Paper I** contained two shorter periods with substantially elevated PM concentrations, during 22 – 24 and 27 – 31 March, presented in the Figure 6.

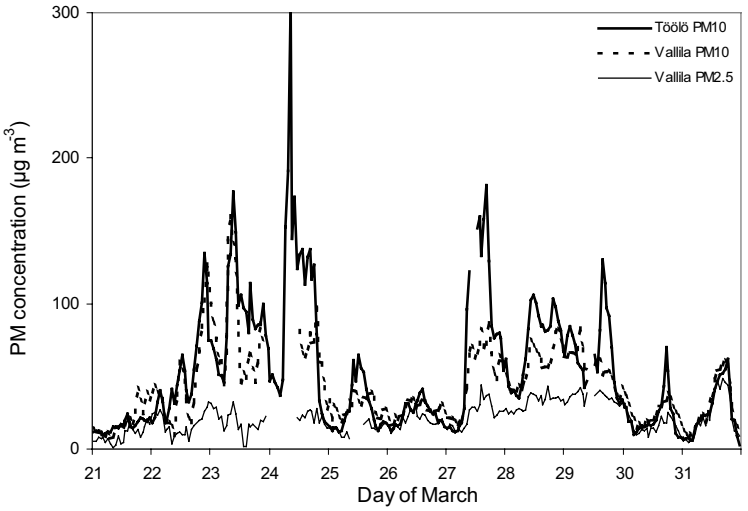


Figure 6. The evolution of the PM_{10} concentrations at the stations at Töölö and Vallila, and the $PM_{2.5}$ concentration evolution at the station of Vallila in the course of the air pollution episode during 21 – 31 March 1998.

An analysis of the measured meteorological parameters (wind speed, relative humidity, surface level atmospheric pressure and temperature) and an atmospheric stability parameter indicated that both of these periods of high concentrations were clearly related to conditions of high atmospheric pressure and relatively low ambient temperatures. The PM concentrations during both 22 – 24 and 27 – 31 March were elevated due to poor mixing produced by low wind speeds in predominantly stable atmospheric conditions.

The polluted air masses could have partly been of long-range transported origin especially on 31 March as the $PM_{2.5}$ concentrations on this particular day were almost identical to the corresponding PM_{10} concentrations at both monitoring stations considered; a detailed analysis would have required air parcel trajectory analyses.

4.3.2 December 1995 Episode in the HMA

An air pollution episode that took place during 27 to 29 December 1995 was studied in **Papers IV** and **V** to obtain information about the meteorological factors leading to elevated air pollutant concentrations and also to examine the performance of two operational versions of the numerical weather prediction model in predicting the meteorological conditions during this air pollution episode.

The evolution of the measured air pollutant concentrations at an urban traffic type site located in the HMA is presented in Figure 7. Concentrations and synoptic, meso- and microscale meteorological conditions, especially vertical temperature profiles, were studied.

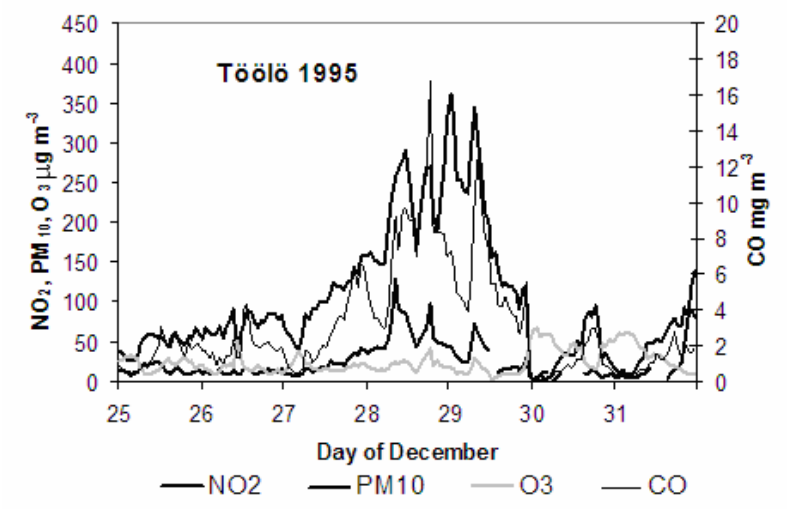


Figure 7. The evolution of the NO_2, O_3, CO and PM_{10} concentrations in the course of the air pollution episode during 25 – 31 December, 1995, at the station of Töölö.

For PM_{10} concentrations, the EU limit value is 50 µg m^{-3} over the averaging period of 24 hours (allowing 35 exceedings per year). The numerical limit value was exceeded at the station of Töölö on 28 December; the 24 hour average value was 58,6 µg m^{-3}. The highest hourly average PM_{10} concentration of 128 µg m^{-3} was measured on 28 December at 8:00 at

the station of Töölö. However, the limit values were not exceeded at the regional background station of Luukki; the comparison of measured regional background concentrations with those measured at urban and suburban stations showed that in this case the increased concentrations were predominantly originated from local sources.

The formation of an extremely strong ground-based radiation inversion in the Helsinki Metropolitan Area was found to have led to the mixing height available for the dispersion of pollutants being exceptionally low. At 00 UTC on 28 December at Kivenlahti, the measured temperature, presented in Figures 8 and 9, increased 15 °C within the lowest 91 m of the atmosphere; at Jokioinen, the corresponding temperature increase was 18 °C within the lowest 120 m. Inversions of such strength are exceptional in southern Finland (Karppinen et al., 2001, Huovila et al., 1991), so this study can be seen as an indicative "worst case" scenario with particular set of weather conditions possible in the area. Inversions of such a temperature gradient have not been reported previously in any other urban areas in Europe (Piringer and Kukkonen, 2002); this is caused by the specific boreal climatological characteristics.

From 27 to 29 December, the wind at the height of 10 m as measured at Helsinki-Vantaa airport was very light. The wind speed varied between 1 and 2 m s^{-1}, being almost calm on 28 December. At this time the upper-level winds were also weak, at least up to 100 m level of the Kivenlahti mast.

Concerning the forecasting of air pollution episodes in the Helsinki Metropolitan Area, the performance of the meteorological models included in the modelling system is important. As the wintertime episodes in the HMA are usually radiation inversion originated (Huovila et al., 1991, Karppinen et al., 2001) the accuracy in forecasting air pollution episodes therefore depends crucially on the ability of the numerical weather prediction models to forecast the strength of these inversions. Examples of measured and corresponding modelled temperature profiles for are presented in Figures 8 and 9.

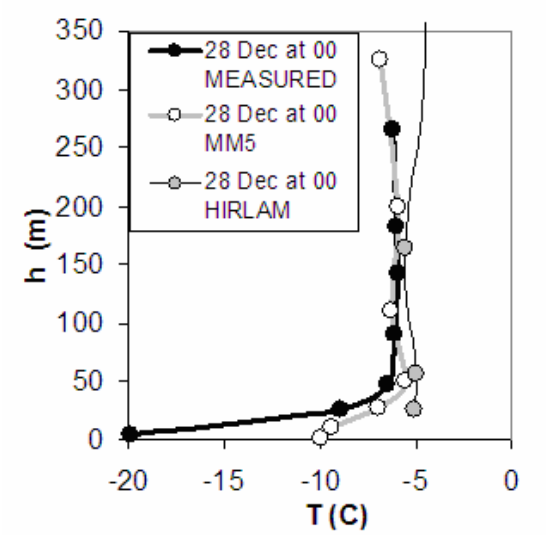


Figure 8. The temperature profiles measured at the station of Kivenlahti, and those predicted by the MM5 and HIRLAM models, 28th December 1995 at 00 UTC.

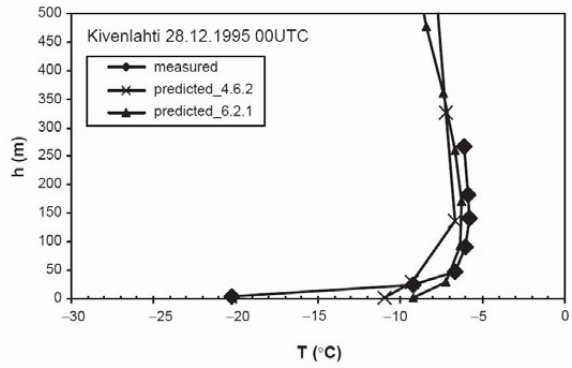


Figure 9. Predicted and measured vertical temperature profiles on 28 December 1995 at 00 UTC at the Kivenlahti meteorological mast. The 24h predictions were made with the HIRLAM versions 4.6.2 (predicted_4.6.2) and 6.2.1 (predicted_6.2.1).

The inversion strengths (°C), reported in the **Paper IV,** predicted by the HIRLAM model version 4.6.2, were substantially weaker or non-existent, as compared both with the corresponding values extracted from the mast data and the sounding data. The HIRLAM model also overpredicted surface temperatures and the height of the inversion layer. A

detailed analysis shows that this under-prediction of inversions is related especially to an over-prediction of ground surface temperatures and near-surface wind velocities, partly caused by deficiencies in the mathematical description of humidity and the state of the ground surface, e.g., the existence of snow cover.

The performance of two versions of the HIRLAM model in operational use were studied closer in **Paper V.** During the episode, both of the versions 4.6.2 and 6.2.1 of the HIRLAM model tended to overpredict wind velocities for the 10 and 30m heights. The less severe inversion on 29 December was fairly well predicted by both models; however, on 28 December, presented in Figure 9, both models substantially overpredicted the low ground surface temperatures.

An overprediction of the inversion height (or the inability of the model to predict the existence of an inversion) would lead to an overestimation of the mixing height. In such a case, pollutant concentrations would be underestimated by any dispersion modelling system utilizing the meteorological data from the HIRLAM model.

So, both of the HIRLAM versions 4.6.2 and 6.2.1 were able to predict the occurrence of the inversions, but the surface temperatures and wind speeds near the surface were generally overestimated, thus leading to underestimation of the strength of the inversion. However, the more recent model version performed better in predicting the ground-based inversions. Sensitivity analyses showed that in episodic conditions, the processes to be modeled can be chaotic, i.e., fairly small variations of input data (or model parameterisations) can lead to major changes in the model predictions.

Berge et al. (2002a) have evaluated the performance of numerical weather forecasting models HIRLAM10 and MM5 in forecasting the weather conditions during one day in Jan 2001 in Oslo, Norway, and the same 27-28 December 1995 episode in Helsinki that is presented here. MM5 was found to predict the height of the inversion quite well, but underestimated the inversion strength in the Helsinki case like in our study.

4.3.3 Selected air pollution episodes in four European cities

In Northern Europe and also in some mountainous areas elsewhere in Europe, strong surface inversions are a key factor in terms of the occurrence of peak pollution episodes. The episodes chosen for closer meteorological analysis in this work were recent and suspected to be at least partly caused by local emission sources in interaction with the local weather patterns. Table 5. contains a summary of the episodes chosen for closer meteorological inspection in this work.

Table 5. A summary of the periods with substantially elevated PM_{10} concentrations during the four selected episodes.

City	Year	Dates	Duration of period
Oslo	2003	4 to 5 January 7 to 10 January	2 days 4 days
Helsinki	2002	3 of April 8 to 13 April	1 day 6 days
London	2003	18 to 27 February	10 days
Milan	1998	14 to 19 December	6 days

As presented in the summary in the Table 6, all the episodes considered were associated with the influence of areas of high pressure (Oslo, Helsinki and London) or of a high pressure ridge (Milan). High atmospheric pressure is commonly related to stable stratification; clearly, however, it does not necessarily lead to extremely stable conditions or strong inversions near the ground level. Inversions can be caused by various atmospheric processes, such as subsidence, fronts, radiation and advection; radiation and advection inversions are those that occur most frequently in the course of air pollution episodes. In the cases examined here, the inversions in Oslo and Milan were mainly caused by advection, and that in Helsinki was a radiation inversion.

Table 6. A summary of main findings for the episodes considered. LRT=long range transport.

City	Character-istic synoptic meteorologi-cal conditions	Characteristic local scale meteorological factors	Type of temperature inversion	Main source category for particulate matter mass	Classifi-cation of the episode
Oslo	An area of high pressure	Temperature inversion, low wind speed, stable stratification	Ground-based, advection, (max 15 °C)	Local wood combustion	Wintertime inversion-induced episode
Helsinki	An area of high pressure	Temperature inversion, low wind speed, stable stratification	Ground-based, radiation, (max 8 °C/50 m)	Suspended dust and local vehicular emissions	Spring dust episode
London	Varying high and low pressure areas	Slight temperature inversion, varying wind speed	Ground-based (less than 1 °C/50 m)	Local vehicular emissions, suspended dust, LRT	Partly local, partly LRT episode
Milan	High pressure ridge	Temperature inversion, Stable stratification	Elevated, advection (max 15 °C/1500 m)	Local vehicular emissions	Wintertime inversion-induced episode

In order to find out whether temperature inversions would also be concomitant with the highest concentrations of PM_{10} in other corresponding episodic cases, episodes contained in the FUMAPEX project database (http://fumapex.dmi.dk) were also investigated. The result found was that in all the corresponding episodes included in the inventory, in Oslo (three local wood combustion-originated cases and two spring dust cases), Helsinki (two spring dust cases) and Milan (two wintertime inversion-induced cases), and London (two cases), this was indeed the case.

In the cases studied here, the best meteorological predictors for the elevated concentrations of PM_{10} were the temporal (hourly) evolutions of temperature inversions, atmospheric stability and in some cases, wind speed. However, in some cases a high wind speed can also increase PM_{10} concentrations, due to an increased suspension of particles from street and ground surfaces. It was also discovered that a low wind speed is not necessarily a good indicator of poor air quality in all locations.

5. Review of the papers

This thesis describes the PM_{10} and $PM_{2,5}$ concentrations measured in the Helsinki Metropolitan Area both on annual time-scale and during specific air pollution episode situations, and analysis and modelling aspects of the spatial and temporal variation of concentrations and temporal variation of the particle population properties. It also compares typical local source air pollution episodes in different types of European urban environments.

Paper I examines the PM_{10} and $PM_{2,5}$ concentration data measured in the Helsinki Metropolitan Area during the years 1997-1999, considering the spatial variation and the temporal variation of the concentrations. Diurnal variation of PM_{10} concentration was found to be clear irrespective of year and the season of the year, and to correspond to a degree with the variation of local vehicular traffic flows. This indicates that the PM_{10} concentrations are originated from vehicular traffic (suspension and direct emissions). For the $PM_{2,5}$ concentrations, both the spatial and temporal variation was found to be moderate, indicating the importance of regionally and long-range transported portion. A wintertime air pollution episode was also described and found to be related to a high pressure system creating low wind speeds and predominantly stable atmospheric stratification which inhibited the dispersion of the pollutants. A long-range transported faction may have been present in the $PM_{2,5}$ concentrations.

Paper II describes the study of the relative importance of different aerosol processes and dilution on particle number concentration and chemical composition in a vehicular exhaust plume in an urban environment, conducted by means of an aerosol process model combined with a simple plume model for dilution. The number concentrations of particles originating from street traffic exhaust underwent a substantial change during the first 25 s after being emitted from the vehicle tailpipe. The concentration of condensable organic vapor was found to have a threshold value below which condensation has no importance, and overall the effects of condensation and coagulation on total number concentrations were shown to be negligible compared to the effect of dilution with urban background air. *The masses given in Table 2 in the Paper II for the background particle composition are too small by*

factors of 10 and 100 compared to the actual mass making up the number concentrations in the two modes. The effect of this can be seen most clearly in Figure 4 c-d) of the Paper II, where the radius of the Aitken mode particles decreases with the simulation time.

Paper III contains corrected results to **Paper II**. The values of particle masses in the urban background air that were used and reported in the **Paper II** for the numerical computations were too small in relation to the corresponding particle number concentrations of urban background air. The main conclusions drawn in **Paper II** are shown to remain valid after the recalculations.

Paper IV describes the course of a wintertime air pollution episode in an urban environment and examines the meteorological situation and other factors affecting the PM, NO_x and CO levels during the episode. Measurement data, and predicted data from the HIRLAM and MM5 weather prediction models was intercompared and used to analyse the episode. An exceptional ground-based temperature inversion leading to exceptionally low mixing height was found to be related to the elevated pollutant levels. An urban dispersion modelling system was also used to predict the pollutant concentrations during the episode.

Paper V evaluates two versions of the numerical weather prediction model HIRLAM against data measured during the air pollution episode described in the **Paper IV**. Both model versions were found to underpredict inversion magnitudes, and they overpredicted nocturnal ground surface temperatures and wind speed close to the ground. These tendencies would lead to overprediction of pollutant dispersion and subsequent underprediction of pollutant concentration levels, if HIRLAM model data would be used in air pollution modelling.

Paper VI presents analysis of PM_{10} (and $PM_{2,5}$ where available) episodes characteristic of four European cities (Oslo, Helsinki, Milan and London). The episodes were chosen so as to be caused mainly by local emission sources. The meteorological data used in the analyses was obtained from measurements, numerical weather prediction models and meteorological pre-processors. The best meteorological predicting variables for the

elevated concentrations were found to be the temporal evolution of temperature inversions, atmospheric stability and wind speed. Low wind speeds lead to less effective mixing and dispersion of pollutants in the air, but also high wind speed can elevate PM concentrations by causing suspension of particles from surfaces.

6. Conclusions

The general aim of this thesis was to evaluate the ambient PM concentrations in the Helsinki Metropolitan Area (HMA), and the factors affecting and leading up to them. This was achieved by a combination of smaller subtasks.

Specifically, the subtasks of this thesis were:

- *to evaluate the sources and temporal variation and to a lesser degree, the spatial variation, of PM in the HMA*.

The diurnal variation of the PM_{10} concentrations in the Helsinki Metropolitan Area measurement sites was shown to be clear, irrespective of the year and the season of the year. This variation partly follows the corresponding variation of local vehicular traffic flows. During the studied years the PM_{10} concentrations were originated mainly from local vehicular traffic (direct emissions and suspension), while the $PM_{2.5}$ concentrations were mostly of regionally and long-range transported origin. A spatial variation of PM_{10} was also shown to exist depending on the distance from traffic sources and their respective traffic volumes.

- *to examine the importance of aerosol processes for the PM concentrations near a traffic source* :

The effects of coagulation, condensation and plume dilution on the evolution of the assumed vehicular exhaust aerosol, concerning particle number concentrations, radii and chemical composition, were considered separately for each particle size mode in a street scale. It was shown that the number concentrations of particles originating from street

traffic exhaust undergo a substantial change during the first 25 s after being emitted from the vehicle tailpipe. Considering the importance of the aerosol processes studied on total number concentrations the dilution process was shown to dominate, while when looking at modeled size-mode wise number concentrations, minimal effect of both condensation and coagulation was seen in the Aitken mode, but not in the accumulation mode. This study would have benefited from better experimental measurement data (containing better description of the measurement, more data points at different distances from the traffic source, and measurement data on particle number concentrations instead of only mass concentrations), which was not unfortunately available at the time.

- *to examine which meteorological variables can be used to predict the occurrence of elevation of PM in the HMA in the local scale, and to evaluate the performance of current numerical weather forecasting models in the case of air pollution episode situations*:

In this thesis, several air pollution episodes containing significant elevations of PM concentrations have been studied. The included episodes were chosen on the basis of occurrence in either winter or spring, and having at least partly local origin.

The HMA episodes were shown to be linked to conditions of predominantly stable atmospheric conditions with high atmospheric pressure and low wind speeds in conjunction with relatively low ambient temperatures.

Both numerical weather prediction model versions were able to predict the occurrence of the inversions, but the surface temperatures and wind speeds near the surface were generally overestimated, thus leading to underestimation of the strength of the inversion. An overprediction of the inversion height (or the inability of the model to predict the existence of an inversion) would lead to an overestimation of the mixing height. In such a case, pollutant concentrations would be underestimated by any dispersion modelling system utilizing the meteorological data from the HIRLAM model. However, the more recent model version performed better in predicting the ground-based inversions.

- *to compare typical air pollution episode conditions in some European cities*:

In the four cities and the cases chosen for study in this work, the best meteorological predictors for the elevated concentrations of PM_{10} during particulate matter episodes were the temporal (hourly) evolutions of temperature inversions, atmospheric stability and in some cases, wind speed. However, in some cases a high wind speed can also increase PM_{10} concentrations, due to an increased suspension of particles from street and ground surfaces. It was also discovered that a low wind speed is not necessarily a good indicator of poor air quality in all locations. Therefore it could be said that the relevance of wind speed for elevated PM_{10} concentrations is dependent of geographical location and not universal.

The phenomena not considered in this thesis but certainly affecting the subject matter include emission modelling for mass-wise measured particulate matter concentrations, the modelling of deposition, suspension and nucleation of particles in the urban environment. Considering that one of the motivations of the study of particulate matter is its influence on the health of human population exposed to it, exposure modelling e.g. the study of periods spent by people in environments where they are exposed to outdoor and indoor particulate matter concentrations, is also a very important consideration. Exposure modelling is not included in this thesis, however. Regional and long-range transport was also not studied in this thesis. It is also conceivable that particulate matter chemical composition is a variable affecting the effects of inhalation of particulate matter on human health, but chemical composition was considered only briefly.

Concerning the modelling of aerosol processes and dilution by background air, the problems in obtaining suitable measurement data were noted. Information about particle mass concentrations was more often available than data about measurements of particle number concentrations, and available material on measurements concerning the particles' chemical composition were few and contained information on the chemical composition size distribution only via the scale of particle mass classes of $PM_{2,5}$ and PM_{10}. Also the variability of the dilution technologies used in emission measurements produced

uncertainty in the modelling input values. Vehicle emissions' particle content is also dependent on the ambient temperature, especially in northern environments where below zero Celsius temperatures prevail during a significant part of the year. Higher particle numbers are emitted in lower temperatures because of particle production by nucleation and subsequent growth by condensation (Kittelson, 1998, Palmgren et al, 2003, Gidhagen et al 2004). Also the availability of emission data in the light of the driving speeds affecting the emission profile was scarce; for example Ketzel et al. (2003) reported having located no studies of emission factors under urban driving conditions in 2003, but only ones with driving patterns corresponding to highway conditions with driving speeds of 70-100 km/h, while driving speeds in the HMA are more of the range of 40-50 km/h (speed regulations, congested traffic).

Particle number concentration measurement data at multiple distances from a major roadway site in Helsinki, obtained from a mobile laboratory (Pirjola et al., 2006), has become available since the publication of **Paper II**. Also more specific information about size-dependent exhaust emissions and background air particles' number concentrations and chemical composition has been published. The roadway site exhaust particles' dilution and transformation processes are now being modeled with a combination of a roadside dilution model (CAR-FMI) and the aerosol process model MONO32.

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